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Spin Dynamics of the One-Dimensional J-J' Model and Spin-Peierls Transition in $CuGeO_3$

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Spin dynamics as well as static properties of the one-dimensional J-J' model ($S = 1/2$, $J > 0$ and $0 \leq \alpha = J'/J \leq 0.5$) are studied by the exact diagonalization and the recursion method of finite systems up to 26 sites. Especially, the dynamical structure factor $S(q, \omega)$ is investigated carefully for various values of α. As α increases beyond the gapless-gapful critical value $\alpha_c = 0.2411$, there appear features definitely different from the Heisenberg model but the same with the Majumdar-Ghosh model. Some of these features depend only on the value of α and not on δ: a parameter introduced for the coupling alternation. By comparing these results with a recent inelastic neutron scattering spectrum of an inorganic spin-Peierls compound $CuGeO_3$ [M. Arai *et al.* : Phys. Rev. Lett. **77** (1996) 3649], it is found that the frustration by J' in $CuGeO_3$ is unexpectedly strong ($\alpha = 0.4$-0.45), and at least α must be larger than α_c to some extent. The value of J is evaluated at ~ 180K consistent with other estimations. The coupling alternation is extremely small. This large frustration is a primary origin of the various anomalous properties $CuGeO_3$ possesses. For comparison we refer also to α'-NaV_2O_5.

KEYWORDS: spin dynamics, neutron scattering, spin-Peierls transition, $CuGeO_3$, J-J' model, Heisenberg model, Haldane-Shastry model, Majumdar-Ghosh model, exact diagonalization, recursion method

§1. Introduction

In connection with the quasi spin gap found in the high-T_c superconductors, materials with a spin gap have been intensively studied both by experiment and theory. For a systematic understanding of the spin gap, it is indispensable to investigate thoroughly the properties of basic models. Among such models, the $S = 1/2$ one-dimensional (1D) antiferromagnetic ($J > 0$) Heisenberg model with frustration by the next-nearest-neighbor exchange, which we call the J-J' model,[1)]

$$\mathcal{H} = J \sum_j \boldsymbol{S}_j \cdot \boldsymbol{S}_{j+1} + J' \sum_j \boldsymbol{S}_j \cdot \boldsymbol{S}_{j+2}, \qquad (1.1)$$

has some unique points. It is well-known that this model changes its character in the low-energy excitation from a gapless feature of the Heisenberg model ($\alpha = J'/J = 0$) to a finite-gap excitation like the Majumdar-Ghosh (MG) case ($\alpha = 0.5$) at $\alpha_c = 0.2411$.[2)] At the MG point the degenerate ground states are exactly known as products of the nearest-neighbor singlet wave functions.[1,3)] So far, some static and thermodynamic properties have been elucidated by numerical studies[4)] and low-energy properties by the conformal field theory.[5,6)] However, systematic studies of spin dynamics have been lacking.[7)]

Another important aspect of this model is a close relationship with the first inorganic spin-Peierls (SP) compound $CuGeO_3$.[8)] The origin of the spin gap in $CuGeO_3$ is now established as the lattice dimerization,[9,10)] although it is very small. Some properties below the SP transition temperature $T_{SP} \sim 14$K are common with the organic SP compounds and can be broadly understood by the theory of Cross and Fisher.[11)] On the other hand, there are many unique aspects contradictory to this theory. For instance, phonon softening has not yet been observed,[12)] which has been necessarily discovered for the organic SP compounds as a driving force of the SP transition. Moreover, the thermodynamic properties observed above T_{SP} do not coincide with the results of the simple Heisenberg model.[8)]

Later, it was found that the latter contradiction can be reconciled by introducing frustration due to the next-nearest-neighbor exchange J'.[13,14)] Although many following theoretical studies[15,16,17)] have unanimously confirmed the importance of this frustration to explain the experiments, the ratio of frustration α remains controversial among them. Originally, Riera and Dobry[13)] concluded $\alpha = 0.36$ by fitting the spin susceptibility $\chi(T)$. Meanwhile, Castilla *et al.*[14)] excluded the possibility for $\alpha > \alpha_c$ by a conjecture based on the theory of Cross and Fisher and the dependence on temperature of the gap, and concluded $\alpha = 0.24$, which is a marginal value for the gapless condition. At any rate, the main point of the controversy is how the gap by J' for $\alpha > \alpha_c$ is compatible with the SP transition.

In this paper, first we investigate spin dynamics of the 1D J-J' model without coupling alternation carefully for various values of α ($0 \leq \alpha \leq 0.5$) by the exact diagonalization and the recursion method at zero temperature.[18)] As a result, the dynamical structure factor $S(q, \omega)$ has qualitatively different characters, according as $\alpha < \alpha_c$ or $\alpha > \alpha_c$; for $\alpha \sim \alpha_c$ it resembles that of the Haldane-Shastry (HS) model.

Second, we compare these results with a complete

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spectrum of the inelastic neutron scattering[19] to search proper parameters extensively in the α-δ space, by adding the effect of coupling alternation δ to the J-J' model eq. (1.1). Note that our fit is carried out at low temperatures ($T \lesssim T_{\rm SP}$) in contrast with the previous ones by $\chi(T)$,[13,14] which were done for $T > 55K$. Consequently, we have found that α thus obtained is unexpectedly large ($\alpha = 0.4$-0.45), and that at least α must be somewhat larger than $\alpha_{\rm c}$. This conclusion is drawn from the fact that the experimental spectrum shows some evident features characteristic of the region $\alpha > \alpha_{\rm c}$, which do not appear only by adding δ to eq. (1.1) with $\alpha < \alpha_{\rm c}$.

Once α is specified, the value of J is determined by neutron data[20] as $\sim$ 180K, which is consistent with those obtained by other means.[13,21] On the other hand, δ is found to be very small (0.001-0.005). We are convinced that the main origin of most unique properties in $CuGeO_3$ is nothing but this large frustration, which is not in competition but in concert with the lattice dimerization in making a gap. In this connection, we refer also to the second inorganic SP compound α'-NaV_2O_5, in which α seems small.

This paper is organized as follows: In §2 we mention the model and the method. Section 3 concentrates on the results for the J-J' model without coupling alternation ($\delta = 0$). In §3.1 the Heisenberg case is taken up as a reference. In §3.2 the regime of $\alpha \sim \alpha_{\rm c}$ is considered in relation to the HS model. In §3.3 characteristics of the gap regime are explained. Section 4 is assigned to the comparison with the experiments of $CuGeO_3$. In §4.1 we compare $S(q,\omega)$ with a complete spectrum of the neutron scattering to determine the value of α. In §4.2 the values of J and δ are considered. In §4.3 based on the parameters thus determined, we discuss a couple of other properties observed in $CuGeO_3$ and α'-NaV_2O_5. We summarize in §5. In Appendix A we describe the single-mode approximation for the J-J' model. In Appendix B we give a proof of a pure single mode at $q = \pi/2$ for the exactly soluble case: $2\alpha + \delta = 1$.

§2. Model and Method

In this section we summarize our model and method briefly. In this paper, we consider the 1D antiferromagnetic Heisenberg Hamiltonian with the next-nearest-neighbor exchange and the coupling alternation:[22]

$$\mathcal{H} = J \sum_j \Big\{ \left[1 - (-1)^j \delta\right] \boldsymbol{S}_j \cdot \boldsymbol{S}_{j+1} + \alpha \boldsymbol{S}_j \cdot \boldsymbol{S}_{j+2} \Big\}, \quad (2.1)$$

where we assume $\alpha, \delta \geq 0$ and $\boldsymbol{S}_{j+N} = \boldsymbol{S}_j$ with N being the number of sites and even.

For $\alpha = \delta = 0$ eq. (2.1) is reduced to the Heisenberg model, for which various ground-state and thermodynamic properties are exactly known by the Bethe Ansatz; quite recently the structure of spin excitation at $T = 0$ has been clarified by using quantum group, etc.[23] The ground-state phase diagram of eq. (2.1) in the α-δ plane was given by the conformal field theory;[5,6] the regime of gapless excitation is limited on the segment $0 \leq \alpha \leq \alpha_{\rm c}$ and $\delta = 0$. At $\alpha_{\rm c}$ marginally irrelevant operators vanish (conformal invariant), so that there exists no logarithmic correction. The critical value was determined as ~ 0.2411[2] by using the fact that the lowest singlet and triplet excited levels cross at this point in the thermodynamic limit. For $\alpha_{\rm c} \leq \alpha \leq 0.5$ and $\delta = 0$ an excitation gap opens as in Fig. 11(b), and the ground states are singlet and twofold degenerate in the thermodynamic limit. But this degeneracy is lifted for finite N and $\alpha \neq 0.5$.

At $\alpha = 0.5$ the degenerate exact ground states are known[1,3] as: $\Psi_\pm = \nu_\pm(\psi_1 \pm \psi_2)$, where

$$\psi_1 = [\ 1,2\ ][\ 3,4\ ] \cdots [N-1, N],$$
$$\psi_2 = [\ 2,3\ ][\ 4,5\ ] \cdots [N, 1], \quad (2.2)$$

and

$$\nu_\pm = 1 \Big/ \sqrt{2^{N/2+1} \pm (-1)^{N/2} 4}\ .$$

Here, the square bracket indicates the singlet pair spin function:

$$[\ i,j\ ] = \alpha(i)\beta(j) - \beta(i)\alpha(j). \quad (2.3)$$

Ψ_+ (Ψ_-) belongs to $Q = 0$ (π).[24] The spin excitations for the MG point were studied by introducing an variational wave function,[3] in which an isolated doublet (or spinon) pair plays an important role.

For other values of α in the J-J' model eq. (1.1), the ground-state and thermodynamic properties are studied by the exact diagonalization and a Quantum Monte Carlo method.[4]

For a finite value of δ, a finite gap always exists. The exact ground state is known on the line $2\alpha + \delta = 1$.[3] In this case, ψ_1, in which singlet pairs sit on the bonds with the larger exchange $J(1+\delta)$, becomes the unique ground state, and ψ_2 (therefore $\Psi_\pm$) is no longer an eigenstate. For $2\alpha + \delta > 1$ the phase is "spiral". In this paper, we concentrate on the parameter range $2\alpha + \delta \leq 1$ ($\alpha, \delta \geq 0$), and leave the spiral phase for future studies.

For this model, our main concern is the spin dynamical structure factor at zero temperature:

$$S(q,\omega) = \sum_n |\langle \Psi_n | S^z_q | \Psi_0 \rangle|^2 \delta\left(\omega - (E_n - E_0)\right), \quad (2.4)$$

where $\hbar = 1$, Ψ_n (E_n) is the n-th eigenfunction (eigenvalue) of the system, and the ground state is indicated by $n = 0$. n runs over all the eigenstates. $S(q,\omega)$ is a quantity proportional to the spin contribution in the intensity of an inelastic neutron scattering spectrum. Since here the ground state is always singlet for finite systems, only triplet states contribute to $S(q,\omega)$ due to the selection rule. For finite systems, $S(q,\omega)$ can be estimated by the recursion (or continued-fraction) method.[18] To this end, we expand the corresponding Green's function $G(z)$ as

$$G(z) = \langle \Psi_0 | S^z_{-q} \frac{1}{z - \mathcal{H}} S^z_q | \Psi_0 \rangle = S(q) C(z), \quad (2.5)$$

where $S(q) = \langle \Psi_0 | S^z_{-q} S^z_q | \Psi_0 \rangle$ and

$$C(z) = \cfrac{1}{z - a_0 - \cfrac{b_1^2}{z - a_1 - \cfrac{b_2^2}{z - a_2 \cdots}}}. \quad (2.6)$$

For finite systems, $C(z)$ is comprised of a finite number of δ functions. Given the exact ground-state wave function, which we usually obtain by the exact diagonalization, we can determine the coefficients a_i's and b_i's through a Lanczos-like procedure. Using this expansion, we can rewrite eq. (2.4) as a sum of residues,

$$S(q,\omega) = -\frac{1}{\pi}\lim_{\eta\to 0}\mathrm{Im}G(\omega+i\eta-E_0) = S(q)\mathrm{Re}\sum_{\ell}\mathrm{Res}\Big[C(\omega+i\eta-E_0),E_\ell\Big], \quad (2.7)$$

where ℓ runs over all the poles of C.

Here, we write down some technical points. Actually, we calculate the contribution to $S(q,\omega)$ by integrating C for each pole with finite η, typically 1-5 $\times 10^{-5}J$. Since the sum rule

$$S(q) = \int_0^\infty d\omega\ S(q,\omega), \quad (2.8)$$

holds, the total contribution of residues for a fixed value of q is normalized to unity irrespective of the value of η. We have checked that the total contribution in our calculation is always $\gtrsim 0.9999$. Next, to raise precision, it is important to start with a ground-state vector as precise as possible. Otherwise, numerical errors are rapidly mingled in the coefficients. In fact, a_i's and b_i's accurate enough to be used seem to be limited to $i \lesssim$ 20-30 at most, so that we cut off higher coefficients than $i = 30$ mostly in this study. This hardly affects poles and residues in low energy and with strong intensity, but causes inaccuracy to some extent of the ones in high energy and with weak intensity. In this context, however, the results are justified for most cases by confirming the regularity of the dependence on system size.

Finally, we mention the finite-size scaling. For the J-J' model, there is no simple scaling function which fits finite-size data correctly.[2] However, by searching for a pertinent function case by case, one can fulfill an extrapolation reliable enough to discuss the physics. We use the usual polynomial fit of order smaller than 10th, in addition to the formula for $\delta \neq 0$:[17]

$$\Gamma(N) = \Gamma(\infty) + \frac{\gamma}{N^m}\exp\left(-\frac{N}{N_0}\right), \quad (2.9)$$

where γ and N_0 are fitting parameters, and $m = 2$ (1) is used if Γ is energy (a quantity of energy difference like the gap Δ).

§3. Spin Dynamics of the J-J' Model

In this section, we focus on the J-J' model eq. (1.1) without coupling alternation. By contrasting these results with the finite-δ cases, the effect of α is definitely discriminated from that of δ.

3.1 Excitation spectrum of the Heisenberg model

Although the spin excitation of the 1D Heisenberg model has been studied for a long period, it was quite recently that the accurate behavior of the spectrum was clarified. In contradiction to the spin-wave picture (spin 1), the excitation spectrum of the Heisenberg model consists of even number of free-spinon (spin 1/2) excitations.[25] As the lowest-order contribution, two-spinon excitations form a dominant continuum, the boundaries of which coincide with the known curves:[26]

$$\omega_\ell(q) = \frac{\pi J}{2}\sin q, \quad \text{and} \quad \omega_{\mathrm{u}}(q) = \pi J \sin\frac{q}{2}. \quad (3.1)$$

We call this continuum as the two-spinon continuum (TSC) in this paper. Some years ago, an approximate formula of $S(q,\omega)$ for the TSC was proposed by Müller *et al.* from the consideration of the exactly soluble XY model, finite-size results and various sum rules, as:[27]

$$S(q,\omega) = \frac{A}{\sqrt{\omega^2-\omega_\ell^2}}\Theta(\omega-\omega_\ell)\Theta(\omega_{\mathrm{u}}-\omega). \quad (3.2)$$

Here, A is a constant of order 1, and Θ is the step function. Actually, the intensity of this spectrum was roughly reproduced by the neutron scattering experiments of $KCuF_3$,[28] a good 1D antiferromagnet. On the other hand, they simultaneously pointed out that it deviates slightly from the exact one mainly due to higher-energy excitations outside the TSC.

Recently, the two-spinon and higher-order-spinon (HOS) contributions to $S(q,\omega)$ have been exactly calculated by using formulae of quantum group,[23] and compared with the exact diagonalization as well as the above approximation.[29] According to these studies, although the formula eq. (3.2) approximately represents the exact two-spinon spectrum, eq. (3.2) misses the behavior in the vicinity of both boundaries. Especially, it overestimates near the upper boundary, under which there has to be a square root singularity in $S(q,\omega)$, instead of a jump given by eq. (3.2). Meanwhile, the two-spinon contribution to $S(q,\omega)$ occupies 72.89% of the total weight; the HOS processes have not a little effect on $S(q,\omega)$ as expected before by Müller *et al.*

Here, we summarize the characteristics of the spectrum of the Heisenberg model as a reference for later discussions, based on our calculations by the recursion method. We note that there have been a number of studies in this line for the Heisenberg model. In Fig. 1 $S(q,\omega)$ of the Heisenberg model is shown for $N = 26$.

Poles by the two-spinon processes have dominant intensity and are situated on a series of sinusoidal curves in the TSC.[27] The characteristics of the TSC are **[a]** the intensity becomes strong as q approaches π due to the logarithmic divergence of $S(q)$ at $q = \pi$. **[b]** At every fixed value of q, $S(q,\omega)$ is monotonically decreasing function of ω.

On the other hand, the other poles are of the HOS contribution, which continuously spread inside ($q/\pi \gtrsim 0.4$) as well as outside of the TSC. The existence of this contribution was suggested[30, 27] in relation to the single-mode approximation as discussed in Appendix A. The intensity of these poles is considerably weak in comparison with the two-spinon contribution, but severely dependent on N and abruptly increases, as seen in Fig. 3(a). The lower boundary of this HOS continuum is indicated by dashed lines; it seems the lowest mode switches a couple of times, and this boundary lowers with increasing N. Incidentally, this curve looks similar to that of the lowest quintet ($S = 2$) excited levels (solid dia-

mond for $N = 14$), which are described by four-spinon processes.[31] The four-spinon continuums of the triplet states and of the quintet states may coincide in the thermodynamic limit, just as the TSC's of the triplet states and of the singlet ones do.[31] At any rate, another important feature in the spectrum of the Heisenberg model is **[c]** there is non-two-spinon contribution, although its intensity is weak.

In the remainder of this subsection, we consider the dependence on system size of the poles and their residues to discuss whether a pole belongs to a continuum or forms an isolated branch. Such a finite-size analysis was successfully used for the detection of an isolated branch in the $S = 1$ Heisenberg model,[32] although we cannot always draw a definite conclusion from this analysis.

Following this work, we plot residues of the poles belonging to the lowest two two-spinon branches in Fig. 2(a). Since both branches belong to a continuum, the value of each residue ought to vanish in the thermodynamic limit. As for the branch 1', the weight decreases with increasing N.[32] However, the weight of the branch 2' is almost independent of N for large q and rather increases with increasing N for medium q within these system sizes. In fact, we have checked various cases and found that the dependence on N of residues is sometimes not monotonic.

On the other hand, Fig. 2(b) shows the dependence on N of the positions of poles for the lowest four two-spinon branches. Although the branch 1' is scarcely dependent on N, other branches have considerable dependence on N. In the inset of Fig. 2(b) the pole positions at $q = \pi$ are plotted versus $1/N^2$. All the branches monotonically decrease as the system becomes large, and seem to gather at $\omega = 0$. Note that $S(q, \omega)$ diverges as $1/\omega$ at $q = \pi$.

As seen in this case, a pole which belongs to a continuum tends to have appreciable dependence on system size at least either of its position or of its residue. On the other hand, if a pole has little dependence on N both of its position and of its residue, and unless adjacent poles converges to its position, that pole probably forms an isolated branch. However, as will be discussed in §4.3, this will not be a necessary condition for an isolated branch. Thus, one should check carefully the dependence on N of the positions and residues in each case.

3.2 Excitation spectrum near the critical point α_c

As mentioned in §2, at $\alpha_c = 0.2411$ the system eq. (1.1) is conformal invariant. This fact reminds us of another conformal invariant model, the HS model:[33]

$$\mathcal{H} = J \sum_{i<j} \frac{1}{d(|r_i - r_j|)^2} \boldsymbol{S}_i \cdot \boldsymbol{S}_j, \tag{3.3}$$

where $d(r) = (N/\pi)\sin(\pi r/N)$. This model is exactly soluble, and not only ground-state but thermodynamic[34] and dynamical[35] properties have been known. $S(q, \omega)$ consists only of the two-spinon processes in contrast to the Heisenberg model, and is given by a formula analogous to eq. (3.2), as:

$$S(q, \omega) = \frac{1}{4} \frac{\Theta(\omega - \tilde{\omega}_{\ell+})\Theta(\omega - \tilde{\omega}_{\ell-})\Theta(\tilde{\omega}_{\rm u} - \omega)}{\sqrt{(\omega - \tilde{\omega}_{\ell+})(\omega - \tilde{\omega}_{\ell-})}}, \tag{3.4}$$

where the boundaries of the continuum are given by $\tilde{\omega}_{\ell-}(q) = Jq(\pi - q)/2$, $\tilde{\omega}_{\ell+}(q) = J(q - \pi)(2\pi - q)/2$ and $\tilde{\omega}_{\rm u}(q) = Jq(2\pi - q)/4$.

With these in mind, let us look at the case of $\alpha \lesssim \alpha_c$. Even if α is switched on, $S(q, \omega)$ basically keeps the feature of the Heisenberg model within the gapless regime. As for the HOS processes, however, their total contribution considerably decreases from the value of the Heisenberg model as shown in Fig. 3(a), which shows the sum of residues by the HOS processes for small values of q/π. In Fig. 3(b), the maximum values of the above sum between $0 \le q/\pi \le 0.5$ are plotted as a function of α for $N = 26$. For $\alpha \sim 0.2$ the HOS contribution becomes minimal.

In view of the conformal field theory, namely as for low-energy processes, these two conformal invariant models are related to each other as follows. If we cut off the exchange coupling longer than the second neighbor in the HS model, the residual part becomes the J-J' model with $\alpha = 0.25$. The marginally irrelevant operators in the discarded long range part cancel out with the corresponding ones in the second neighbor coupling of $0.25 - \alpha_c$. Thus, it is natural to expect that the behavior of $S(q, \omega)$ for small ω at $\alpha \sim \alpha_c$ resembles that of the HS model. Strictly speaking, however, the HOS contribution never vanishes and the minimum contribution is not at α_c but ~ 0.18-0.2. This means that the behavior of higher-energy processes deviates to some extent from the expectation in the infrared limit.

Such tendency can be seen in Fig. 4, which shows $S(q, \omega)$ for $\alpha = 0.2$. The shape and the size of the main continuum almost coincide with those of the HS model; at the upper edge of the continuum, $S(q, \omega)$ is more likely to have a jump than that in Fig. 1. For more quantitative discussions on $S(q, \omega)$ the introduction of the n-th frequency moment defined as,

$$K^{(n)}(q) = \int_0^\infty d\omega\ \omega^n S(q, \omega). \tag{3.5}$$

is convenient. First, we consider static susceptibility $\chi(q) = K^{(-1)}(q)$, which is depicted for some values of α in Figs. 5(a) and 5(b).

For the Heisenberg model, the uniform susceptibility $\chi(0)$ is given by the Bethe Ansatz;[36] we choose the normalization as $\chi(0)J = 1/(2\pi^2)$. For a general value of q, diagonalization results have been discussed recently;[29] $\chi(q)$ is expected to diverges as $1/(\pi - q)$ with some logarithmic correction. What we should add here is that as in the inset of Fig. 5(a) slight dependence on N is observed for small q, and that the extrapolated value for $q = 0$ seems to deviate subtly upward from Griffiths' value. This may possibly means the existence of some logarithmic singularity in the small-q limit owing to a non-fixed-point model.

As α increases, $\chi(q)$ increases for medium values of q. The tendency becomes opposite for the higher-order frequency moments, as will be mentioned shortly. As discussed above, the curve for $\alpha = 0.2$, rather than α_c, coincides well with that of the HS model except for $q \sim 0$, where the long-range part of the coupling probably plays a major role. By the way, $\chi(q)$ for the HS model is

obtained by integrating eq. (3.4) as,

$$\tilde{\chi}(q)J = \frac{1}{2}\frac{1}{\sqrt{|\tilde{\omega}_{\ell-}\tilde{\omega}_{\ell+}|}}\left[\arcsin\left(\frac{q}{\pi}-1\right)+\frac{\pi}{2}\right]. \quad (3.6)$$

In contrast with the Heisenberg case, the leading power is linear in q for $q \to 0$ and $\tilde{\chi}(q) \propto 1/(\pi - q)$ without logarithmic correction for $q \to \pi$.[37]

We discuss the cases of $\alpha > \alpha_c$ together here. Since the system has a finite gap, $\chi(q)$ vanishes for $q \to 0$. As a sign of it, the abrupt decrease of $\chi(q)$ can be seen for large values of α. The value of q at which $\chi(q)$ drops corresponds to π/ξ, where ξ is the spin correlation length. For the MG case ($\alpha = 0.5$), the spin correlation S_i vanishes for $i \geq 2$; the anomaly exists at $q \sim \pi$. By extrapolation we obtain $\chi(\pi)J \sim 0.843$.

Leaving the discussion about $K^{(0)}(q) = S(q)$ and $K^{(1)}(q)$ for §3.3 and Appendix A respectively, here we consider $K^{(2)}(q)$ and $K^{(3)}(q)$. As the order n of the frequency moment becomes higher, the effect of high-energy processes is amplified. In Figs. 6(a) and 6(b), the second and third frequency moments are plotted, respectively. As n increases, the values of the J-J' model for $q \sim \pi$ are enhanced in comparison with that of the HS model given by the integration of eq. (3.5):

$$\frac{\tilde{K}^{(2)}(q)}{J^2} = -\frac{q}{128}(2\pi - q)(7q^2 - 14\pi q + 6\pi^2) - \frac{1}{32}(q-\pi)^2(2q^2 - 4\pi q + 3\pi^2)\ln\left(1-\frac{q}{\pi}\right), \quad (3.7)$$

$$\frac{\tilde{K}^{(3)}(q)}{J^3} = \frac{q}{768}(2\pi - q) \times (28q^4 - 112\pi q^3 + 169\pi^2 q^2 - 114\pi^3 q + 30\pi^4) + \frac{1}{64}(q-\pi)^4(2q^2 - 4\pi q + 5\pi^2)\ln\left(1-\frac{q}{\pi}\right). \quad (3.8)$$

A main cause is probably the HOS contribution of the J-J' model outside the TSC, as pointed out for the Heisenberg case.[29] The enhancement for small q and large α is owing to the considerable intensity beyond the upper edge of the main continuum in the gap regime as in Fig. 3(b). This is one of the topics in §3.3.

Finally, we point out one more important fact that the height of the TSC becomes shorter monotonically with increasing α. The highest point of the TSC continuum (at $q = \pi$) is πJ for the Heisenberg model, and $\pi^2 J/4$ for the HS model, which is comparable with the case of $\alpha \sim \alpha_c$. The influence of it appears also in Figs. 6(a) and 6(b); as α increases, $K^{(n)}(q)$ for large n generally decreases. We return to this point later when we estimate the value of J by experimental spectra.

Having described mainly the resemblance of $S(q,\omega)$ for $\alpha \sim \alpha_c$ to that of the HS model, we close this subsection by summarizing the properties of $S(q,\omega)$ for $\alpha \lesssim \alpha_c$ (for example, Fig. 4) for later discussions. Basically, the behavior of $S(q,\omega)$ remains having common features with the Heisenberg model; we emphasize repeatedly **[a]** the intensity of $S(q,\omega)$ becomes considerably strong as q approaches π due to the divergence of $S(q)$. **[b]** The intensity of $S(q,\omega)$ is strongest at the lower edge and a monotonically decreasing function of ω for every q. As for the point [c], although non-two-spinon contribution exists, their intensity becomes extremely weak for $\alpha \sim \alpha_c$, where the features of $S(q,\omega)$ resemble that of the HS model.

3.3 Excitation spectrum for $\alpha > \alpha_c$

Since the characteristics of the gap regime are typically realized in the MG case ($\alpha = 0.5$), we take it up first in this subsection. In this case we use $\Psi_\pm$ as the starting vectors in the recursion method. In Fig. 7(a) we show the residues of $C(z)$ [eq. (2.6)] for $N = 24$; we plot both contributions from Ψ_+ ($Q = 0$) and Ψ_- ($Q = \pi$) together, therefore the spectrum is completely symmetric with respect to $q = \pi/2$.[38] In Fig. 7(b) $S(q,\omega)$ is shown for $N = 26$. We itemize their characteristics in comparison with the gapless regime below.

[1] There is appreciable intensity outside the main continuum not only for the small-q region [see also Fig. 3(b)] but in the vicinity of $q = \pi$. On the contrary, there is little high-energy contribution around $q = \pi/2$.

[2] $S(q,\omega)$ is not a monotonically decreasing function of ω; the upper edge of the continuum for $q > \pi/2$ has stronger intensity than the inner area of the continuum.

[3] The region near the lower edge of the continuum has strong weight, like the gapless cases. However, the weight of $S(q,\omega)$ between $q = \pi/2$ and π is comparable, because the residues become huge around $q = \pi/2$, as well as $S(q)$ does not diverge at $q = \pi$.

[4] An isolated branch appears for $0.45 \lesssim q/\pi \lesssim 0.55$. As seen in Fig. 7(a) this isolated branch becomes a pure single mode for $q = \pi/2$ with $\omega = J$ independent of N. The weak intensity in high energy and the strong intensity at the lower edge around $q = \pi/2$ mentioned in [1] and [3] respectively are attributed to this isolated branch.

These features are in sharp contrast with the ones [a]-[c] for $\alpha < \alpha_c$.

Let us look at the spectra more closely. The structure of the continuum is more complicated than the TSC in the gapless regime; near the upper edge it is not easy to trace each branch, while a series of dominant branches can be distinguished in the lower half of the continuum. In particular, near the lower edge, a number of branches with comparable weight seem to gather. The lowest branch does not have the strongest intensity.

To check the dependence on N, we show the residues of the four lowest dominant branches in Figs. 8(a) and 8(b). For $q/\pi \gtrsim 0.55$ the weight of every branch depends severely on system size. Although the branches 1' and 2' monotonically decrease and the branch 4' monotonically increases with increasing N, the branch 3' shows complicated behavior. Positions of the corresponding poles are shown in Fig. 8(c). The higher the energy of the branch is, the severer the dependence is. As shown in the inset of Fig. 8(c), which shows the pole positions versus $1/N^2$ at $q = \pi$, every branch seems to converge to the lower edge.

It is clear that the spectrum forms a continuum for $q/\pi \gtrsim 0.55$, because both the residues and the poles are considerably dependent on system size. On the other hand, as in the Figs. 8(a) and 8(b), for $q/\pi \lesssim 0.55$ the

weight of the branches 1' and 2' is enhanced rapidly as q approaches $\pi/2$ without dependence on N. Inversely, the branches 3' and 4' seem to vanish. As for the pole positions of the branches 1' and 2', there is little dependence on N [Fig. 8(c)]. Consequently, according to the discussion in §3.1, an isolated branch with weight more than 90% seems to occur in this region. In fact, at $q = \pi/2$ it is proven that the spectrum becomes a pure single mode as shown in Appendix B. Incidentally, a similar spectrum was obtained by using a variation method;[3] in addition to a continuum, a bound state below the continuum was derived for $0.36 \lesssim q/\pi \lesssim 0.64$.

Next, we touch on the point [2]. In Fig. 9 the residues at $q = \pi$ are plotted versus ω. Near the lower edge, there exists a large peak like the gapless cases. However, it is not clear in this case whether this peak diverges, and whether it is situated exactly at the lower edge. Anyway, the weight in the continuum once has a broad minimum ($\omega/J \sim$ 1.5-1.8), then another peak appears near the upper edge ($\omega/J \sim 2$). This peak seems less dependent on N, and has tails in both sides of ω. Similar tendency can be seen for other values of q. This profile is in sharp contrast with those in the gapless region.

Having focused on the MG case, here we mention the cases of smaller values of α. In these cases, the ground state is not twofold degenerate except for the thermodynamic limit, thus we treat excitations only from the nondegenerate ground state for finite N. In the thermodynamic limit, the two ground states with $Q = 0$ and π will give contributions mutually symmetric with respect to $q = \pi/2$ just like the MG case.

Figures 10(a)-10(c) show $S(q, \omega)$ for $N = 26$ and $\alpha =$ 0.35, 0.4 and 0.45, respectively. In these figures we can recognize that the characteristics of the MG model [1]-[4] still remain. Comparing these with Fig. 4 ($\alpha = 0.2$) and Fig. 7(b) ($\alpha = 0.5$), we find that the spectrum gradually changes from the gapless feature to the one of the MG case.

Finally, for later discussions, we represent some static quantities; Fig. 11(a) shows the ground-state energy, and Fig. 11(b) the lowest excitation at $q = \pi/2$ and the gap Δ. These values are extrapolated from the finite-size data by the polynomial fit of second and fourth orders. There is no critical behavior at $\alpha = \alpha_c$.

The static structure factor $S(q)$ both for the Heisenberg model[29] and the HS model[39, 35] is logarithmically divergent at $q = \pi$. Accordingly, the situation is basically the same for the gapless regime of the J-J' model. On the other hand, as α increases beyond α_c, the divergence at $q = \pi$ is suppressed. For the MG point $S(q) = (1 - \cos q)/4$, namely $S(q)$ is not only convergent but upward convex near $q = \pi$. In Fig. 12 $S(q)$ for large α is shown. Even for $\alpha = 0.4$, $S(q)$ is considerably enhanced near $q = \pi$. Furthermore, it increases with increasing system size. On the other hand, for $\alpha = 0.45$ the dependence on N of $S(\pi)$ is in the direction of suppression.

Anyway, concerning the excitation in the regime of $\alpha > \alpha_c$, we are still far from understanding in the standard of the Heisenberg model. Further studies are necessary.

§4. Comparison with Spin-Peierls Compounds

Based on the discussion in the preceding section, we consider various aspects of the SP transition in $CuGeO_3$ and α'-NaV_2O_5.

4.1 Complete spectrum of inelastic neutron scattering

Recently, a complete spin excitation spectrum of $CuGeO_3$ was obtained by the inelastic neutron scattering by Arai *et al.*[19] The gap and the size of the continuum in this spectrum are consistent with the previous neutron experiments.[20, 10, 40] At a glance on the spectrum for $T = 10$K (Fig. 1 of ref. 19), it looks like the TSC of the Heisenberg model. However, as the authors pointed out, there are points which are incompatible with the Heisenberg model. We itemize such points below including the authors' words.

[0'] The shape of the lower and the upper edges of the continuum is wedge-like and does not have the roundness characteristic of sine ($\alpha = 0$) and quadratic ($\alpha \sim \alpha_c$) curves. Concerning this point, compare Figs. 18(a) and 19(a), for instance.

[1'] There is appreciable intensity above the continuum, especially for $q \sim \pi$.

[2'] There is "rampart" or ridge of scattering surrounding a valley in the spin continuum.

[3'] There exists a peak in intensity on the lower boundary of the continuum at zone boundaries (0,0,$\pi/2$) and (0,0,$3\pi/2$).

The points [1']-[3'] completely correspond to what we have summarized as [1]-[3] for $\alpha > \alpha_c$ in §3.3, respectively. In this viewpoint, let us compare Fig. 1 of ref. 19 with Figs. 7(b) and 10(a)-10(c) in this paper, before taking account of the effect of coupling alternation. Among the above four panels Fig. 10(c) ($\alpha = 0.45$) seems to bear the best resemblance.

As for the point [1'], the intensity of every pole over the upper edge of the main continuum ($q \sim \pi$) in Fig. 10(c) corresponds to the one in Fig. 1 of ref. 19. For example, a horn-like short branch jutting out upward from $q = \pi$ just above the main continuum ($\omega/J \sim 2.1$) is distinctly reproduced. Incidentally, because the spectrum should be symmetric with respect to $q = \pi$, the asymmetric intensity in the experimental spectrum, especially for $q > \pi$, is due to the contamination by phonons etc., as the authors mentioned.

Concerning the point [2'], the strong intensity at the upper edge and the weak intensity in the upper half of the continuum for $q \sim 2\pi/3$ - π are quite similar between the two figures. Recall the profile in Fig. 9 for the MG case.

As for the point [3'], the experimental intensity near the lower edge is almost the same between $q = \pi/2$ and π. Furthermore, the maximal points seem to be situated slightly above the exact lower edge, which we have suggested in §3.3, although it is not clear whether this is due to its intrinsic property or thermal broadening.

For smaller values of α [Figs. 10(a) and 10(b)], the tendencies deviate from the experimental spectrum. For example, in Fig. 10(a) for $\alpha = 0.35$, the strongest intensity, as to the lower edge, is clearly at $q = \pi$. If

$\alpha \sim \alpha_c$, the intensity over the continuum considerably attenuates, as discussed in §3.2. Consequently, the value of α should be fairly larger than α_c.

More quantitative fit is possible; in Fig. 13 we compare integrated amplitude $S(q)$ (Fig. 12) with the neutron data, which are taken from Fig. 2 of ref. 19. In the experimental results, there is neither divergence nor abrupt increase for $q \sim \pi$ characteristic of smaller values of α. It reads $\alpha \sim 0.45$ for T=10K and $\alpha \sim 0.5$ for $T = 20$K ($T_{SP} = 14$K), which is consistent with the above direct comparison of $S(q,\omega)$. This indicates that the value of α hardly changes through T_{SP}, or reduces slightly for $T < T_{SP}$ if any. Incidentally, there is a tendency of the experimental data to be rather smaller than the diagonalization result for $q \sim \pi$. This is probably due to two facts. One is that in obtaining $S(q)$ by integrating experimental $S(q,\omega)$, the higher-frequency contribution than 32meV (the top of the continuum) is cut off to exclude the contribution by phonon. Actually, there remains magnetic contribution of several percent, as mentioned above. The other is that the introduction of δ somewhat reduces $S(q)$ at $q = \pi$. In this connection, if a smaller value of α is assumed, a considerable large value of δ is needed to reproduce the experiment, for example, $\delta \gtrsim 0.03$ even for $\alpha = 0.4$; such a parameter set is incompatible with the value of Δ, as we will see later.

Thus, we have found that most features of this neutron spectrum are explained by the J-J' model with $\alpha = 0.4$-0.45, even without introducing δ.

Next, let us check whether or not the above aspects are also realized by introducing δ without α or with smaller values of α. Figures 14(a)-14(c) show $S(q,\omega)$'s for $\delta = 0.05$ and three values of α.[41] To emphasize the effect of δ, a fairly large value of δ is used for $CuGeO_3$ (δ is of order 0.001). In these figures some new features can be seen. For example, as will be discussed in §4.3, there appears an opening of intensity between the lowest branch and the above continuum, especially for large α. However, for the case of $\alpha < \alpha_c$ [Figs. 14(a) and 14(b)] the characteristics [1]-[3] for $\alpha > \alpha_c$ discussed in §3.3 do not appear even if δ is added, while these characteristics are preserved for $\alpha > \alpha_c$ if δ is added, as in Fig. 14(c). In other words, the characteristics [1]-[3] appear only when $\alpha > \alpha_c$.[42] Furthermore, these characteristics do not become manifest unless α is fairly larger than α_c. For smaller values of δ, the characteristics [1]-[3] do not appear for $\alpha < \alpha_c$, of course. Thus, these facts exclude at least the possibility of $\alpha < \alpha_c$, which is contrary to the discussion of Castilla *et al.*[14] We will point out a mistake in their assumption in §4.3.

4.2 Estimation of J and δ

First, we discuss the value of J. The first estimation of J was given by a neutron experiment as $J \sim 120$K,[20] in which the maximal point of the lowest branch in the spectrum was compared with $J\pi/2$, which is obtained from the des Cloizeaux-Pearson curve [the first equation of eq. (3.1)] for the Heisenberg model. However, the experimental spectrum has to be compared with that of eq. (2.1). As we have already shown in Fig. 11(b), the size of the continuum decreases with increasing α. The maximal value of the lower edge ($q = \pi/2$) decreases from $J\pi/2$ to J, as α changes from 0 to 0.5. Since $\alpha = 0.4$-0.45, J is modified into a larger value by ∼50%; thus we have $J \sim 180$K.

In the above estimation of J we have neglected the effect of δ; let us check it below. In Fig. 15 the maximal values of the lower edge are plotted as a function of δ for some values of α. In contrast with the effect of α, the spectrum size weakly depends on δ. Although it abruptly increases for small δ and large α, the increment is at largest a few percent, because δ is extremely small. Consequently, the coupling alternation scarcely affects the estimation of J.

Now, we refer to the estimations of J by other means. Many authors tried to evaluate α and J by comparing with $\chi(T)$ by experiment,[8] which shows a broad peak around 56K. Among them, Riera and Dobry obtained $\alpha = 0.36$ and $J = 160$K, by fitting the maximal point of $\chi(T)$.[13] Since this fit was quite good for $T > 56$K, following studies often adopted these values.

Using Faraday rotation, Nojiri *et al.*[21] estimated J at 183K; they compared the field at which magnetization was saturated with the formula for the Heisenberg model: $g\mu_B H_s = 2J$. Although in this case the corresponding value for eq. (2.1) should be used for accuracy, it is convenient to use, instead, the known formula for the J-J' model:[4]

$$g\mu_B H_s/J = \begin{cases} 2 & 0 \le \alpha \le 0.25 \\ 1 + 2\alpha + 1/8\alpha & \alpha \ge 0.25 \end{cases} . \quad (4.1)$$

Unless the variation by δ is singular at $\delta = 0$, this estimation is not poor because δ is very small. As a result, the value is slightly modified as $J = 178$K, 173K, 168K for $\alpha = 0.35$, 0.4, 0.45, respectively.

Thus, the estimation of J by the neutron experiments, $J \sim 180$K, is consistent with other estimations. By using $\alpha = 0.45$ (0.4) and $J = 180$K, the gap energy without coupling alternation is evaluated at 19K (5K), which must be a value too small to make a gap by itself above T_{SP}.

Next, we estimate the coupling alternation parameter δ. The excitation gap was estimated by some experiments. In particular, the neutron scattering experiment can directly observe the gap as a function of temperature $\Delta(T)$, and concluded $\Delta(0) = 2.1$meV (24K),[20] which is consistent with other experiments like NMR,[43] specific heat,[44] magnetic susceptibility $\chi(T)$,[8] etc.

In Figs. 16(a) and 16(b), the gap Δ versus δ obtained by the exact diagonalization for eq. (2.1) is depicted. In Fig. 16(b), the dash-dotted (dashed) line corresponds to $\Delta = 24$K and $J = 180$K (160K). If we assume $\alpha = 0.45$ (0.4) and $J = 180$K, δ becomes as tiny as 0.001 (0.005). This value is much smaller than the previous estimations: $\delta = 0.012$-0.014.[13,17] This difference is originated in the fact that Δ is an abruptly changing function of α for ~ 0.5 and of δ for ~ 0.

Experimentally, the atomic displacement due to the lattice dimerization was measured for $CuGeO_3$ by neutron diffraction.[10] The displacement ratio δ_ℓ is very small; $\delta_\ell \sim 0.001$ both for Cu along the c axis parallel to the 1D chain and for O(2) along the axes perpendic-

ular to the chain. This value is too small to detect in some other means.[45] However, it is still unknown how this small displacement is connected to a small exchange coupling.

In $CuGeO_3$, CuO_6 octahedra link mutually with edge sharing along the c axis; the angle between Cu-O-Cu is 98°. The exchange coupling, which is determined by the overlap of the involved localized orbitals, tends to be weakly ferromagnetic in the case of the right angle, as Kanamori classified many years ago.[46] On the other hand, recently Geertsma and Khomskii argued that for the realization of antiferromagnetic coupling the side-group effect of Ge plays an important role.[47] Thus, we cannot assert the relation $\delta_\ell \propto \delta$ necessarily holds.

By the way, from the above discussion one can expect relatively large long-range exchange couplings survive in edge-sharing materials. Hence, we have checked the third-neighbor's effect for various strength of $J^{(3)}$ (> 0) and for some values of α and δ. The common aspect among the resultant spectra is that the top of the lower edge of the continuum becomes broad and flat around $q = \pi/2$ with the spectral size almost unchanged. This aspect is quite different from the characteristics of $CuGeO_3$. Thus, the third-neighbor exchange is found negligible.

Now, we refer to the second inorganic SP compound α'-NaV_2O_5,[49] in which VO_5 pyramids link mutually with corner sharing in the 1D direction. Since $\chi(T)$ for $T > T_{\rm SP}$ is well fitted by the Bonner-Fisher curve[48] with J =560K, the frustration by J' seems small in this compound. The gap is estimated at $\Delta = 98K$ by NMR.[50] Using these values or another estimation of J = 441K and Δ = 85K,[51] we have δ = 0.043-0.048 from the curve of $\alpha = 0$ in Fig. 16(a).[52] Since the relation $\delta_\ell \propto \delta$ holds in this case, the displacement due to dimerization is probably much larger. In fact, a split in the Na-NMR spin-echo intensity corresponding to the chain direction has been observed below $T_{\rm SP}$,[50] in contrast with $CuGeO_3$.[43, 45]

Before closing this subsection, let us look at the SP transition in the light of energy. We depict the total magnetic energy E/J for $\delta = 0$ and $\delta \neq 0$ in Figs. 11(a) and 17, respectively. As temperature is lowered, the lattice will be distorted so as to minimize E/J. Thus, the displacement at $T = T_{\rm SP}$ will tend to enhance δ as well as J and lower α. This tendency agrees with the one seen in Fig. 13, although the thermal fluctuation may be also involved in this case.

4.3 Further discussions

In this subsection, we will consider a couple of issues in relation to the experiments.

First, we take up $\Delta(T)$. Harris *et al.*[53] discussed this issue by combining three materials: (1) The dependence on temperature of the intensity of a superlattice peak: $I(T) \propto [T_{\rm SP} - T]^{2\beta}$. The work by neutron diffraction [54, 55] and X-ray diffraction[56] concluded $\beta \sim 0.33$ in agreement. (2) The spontaneous contraction along the b axis with the Ginzburg-Landau theory: $I \propto \delta_\ell^2$. (3) The Cross-Fisher theory: $\Delta \propto \delta^{2/3}$. Thus, they obtained $\Delta \propto [T_{\rm SP} - T]^{2a\beta}$ with $a = 1/3$, tacitly assuming $\delta \propto \delta_\ell$.

Later, Castilla *et al.*[14] excluded the possibility of $\alpha > \alpha_{\rm c}$, based on the above discussion and the renormalization-group theory. They assumed $\Delta \propto \delta^{2/3}$ with negligible logarithmic correction throughout the gapless regime, while $\Delta \propto \delta$ for $\alpha > \alpha_{\rm c}$. However, this assumption is false. As in Fig. 16(a), Δ is nearly proportional to δ for small δ and $\alpha = 0$. The leading power $\delta^{2/3}$ is smeared out due to the severe logarithmic correction, even for large values of δ. This aspect had been already pointed out by the diagonalization studies.[57, 31] On the contrary, the leading power $\delta^{2/3}$ was clearly observed for $\alpha = \alpha_{\rm c}$ in the density matrix renormalization group calculations by Chitra *et al.* ,[6] because the model becomes conformal invariant and there is no logarithmic correction, as mentioned in §3.2. Our data for $\alpha = 0.2$ is consistently fitted with a power of ~ 0.71. As α becomes larger, the seeming power further decreases with a finite gap at $\delta = 0$ and approaches 0.5 near the MG point. These results are rather contrary to the assumption by Castilla *et al.* Consequently, there is no theoretical reason that α has to be smaller than $\alpha_{\rm c}$.

Actually, the value a in the power of $\Delta(T)$ was estimated at ~ 0.15,[55] which is much smaller than the seeming power in Fig. 16 of ~ 0.26 for $\alpha = 0.45$, as well as the prediction of the Cross-Fisher theory of 1/3. However, the discussion of power itself is meaningless for the gap, which has serious logarithmic modification.[58] Furthermore, it is not certain whether the relation $\delta_\ell \propto \delta$ holds for edge-sharing materials like $CuGeO_3$, as discussed before. On the other hand, $\Delta(T)$ for α'-NaV_2O_5 also shows a similar abrupt increase at $T = T_{\rm SP}$.[59] These may indicate that $\Delta(T)$ is independent of the details of the exchange coupling.

Second, we consider the double gap reported in a recent neutron experiment for $CuGeO_3$.[60] According to it, gap-like weak intensity was observed above the lowest peak at 2.1meV for $q = \pi$; this "second gap" had a similar width with the first one.

Let us return to Figs. 14(a)-14(c) for $\delta = 0.05$. Comparing these figures, we recognize that the aspects are different between Figs. 14(a) [or 14(b)] and 14(c). In the former case ($\alpha < \alpha_{\rm c}$) the lowest branch for $\delta = 0$ [Figs. 1 and 4] splits into two and the opening between them becomes wider with increasing α; at $q = \pi$, however, these two branches merge and the second gap seems to vanish. Meanwhile, in the latter case ($\alpha > \alpha_{\rm c}$) the second gap is open for all the range of q. Thus, this issue is highly dependent, at least quantitatively, on the value of α and δ. Here, we focus on the plausible parameters for $CuGeO_3$ and α'-NaV_2O_5, leaving more general discussions for future publication.

To begin with, we consider the case of $\alpha = 0.45$, namely $CuGeO_3$. Shown in Figs. 18(a) and 18(b) is the dependence on N of the pole position and the residue respectively corresponding to the two lowest branches for $\delta = 0.01$. We use a somewhat larger, but sufficiently small, value of δ than our estimation so as to find its effect.

First, we discuss the branch 1'. Both the pole position and the residue of the branch 1' scarcely depend on system size except for $q \sim 0$ or π. For $q = \pi$, relatively

reliable extrapolation for both quantities is possible, as shown in the inset of Fig. 18(a) and by an arrow in Fig. 18(b). Moreover, since the pole position of the branch 2' is always away from the branch 1', as will be discussed below, the branch 1' forms an isolated mode.

Next, as for the branch 2' both the pole position and the residue are almost independent of system size for medium values of q, although the behavior of the residue is complex. For $q = \pi$, the pole position converges to a value which is higher than that of the branch 1', as shown in the inset of Fig. 18(a). On the other hand, we are not certain that the residue of the branch 2' at $q = \pi$ has finite weight for $N \to \infty$. Concerning the branch 3' (not shown in the figures), both the pole position and the residue depend on N, but we cannot elicit definite answers from our data whether its pole position converges to that of the branch 2'. Consequently, the branch 2' probably forms another isolated branch, although there remains the possibility of the lower edge of the continuum. The branch 3' belongs to the continuum.

Anyway, the opening between the branches 1' and 2' exists for every value of q. This opening is thin for medium values of q and relatively wide at $q = \pi$. Nevertheless, the width of this second gap at $q = \pi$ is fairly smaller than that of the first gap, as shown in the inset of Fig. 18(a). Incidentally, if the branch 2' is an isolated branch at $q = \pi$ too, the third gap will also exists.

We have confirmed this situation remains the same for some similar values of the parameters. On the other hand, for larger values of δ (for example $\alpha = 0.45$ and $\delta = 0.1$), the branches 1' and 2' form clear isolated modes, as studied formerly.[17] Even in this case, however, the width of the second gap is found still smaller than the first gap. This result is in sharp contrast with an RPA calculation,[61] in which these two gaps have the same width.

Now, we turn to the case of $\alpha = 0$ and $\delta = 0.05$ with α'-NaV_2O_5 in mind.[52] In Fig. 19(a) we show the pole positions of the lowest three branches with dominant intensity. The branches 1' and 2' are almost independent of system size for all the range of q. A conspicuous point is that these two branches merge into a single pole at $q = \pi$. On the other hand, the poles belonging to the branch 3' have severe dependence on system size. Moreover, we find by the extrapolation of the poles at $q = \pi$ that the branch 3' converges to the branch 2' [the inset of Fig. 19(a)]. Shown in Fig. 19(b) are the corresponding residues. As for the branch 1', the dependence on N is seen for $q/\pi \gtrsim 0.6$; the branch 2' also has large dependence on N. Consequently, we can interpret the above as follows: the branch 1' is isolated except for $q \sim \pi$, and the branch 2' is the lower edge of the continuum. Thus the second gap vanishes for $q = \pi$, while an isolated branch appears appreciably below the continuum for q away from $q = \pi$. Furthermore in the opening between them there is weak intensity of the higher-order processes for $q/\pi \sim 0.5 \pm 0.2$. The situation is quite different for larger values of δ ($\gtrsim 0.15$), for which there appears an obvious second gap even at $q = \pi$. However, the second branch belongs to a continuum unlike the cases of large α.

Summarizing, the appearance of a second gap or isolated branches is dependent on the values of α and δ. The second gap, which is narrower than the first gap, may be found at $q = \pi$ for $CuGeO_3$. On the contrary, an isolated branch will be observed for $q \neq \pi$ for α'-NaV_2O_5.

Finally, we mention a connection with the neutron experiments under high pressure.[62] T_{SP} increases linearly with increasing pressure.[63] The lattice constant along the chain (c axis) becomes slightly longer, while the displacement of oxygen in the ab plane is rather large. The gap energy becomes twice larger (4.1meV) under 1.8GPa at 5K. On the other hand, the dimerization for Cu atoms decreases under high pressure.

One possible interpretation of the above experiments, which seem mutually contradictory at a glance, within the model of eq. (2.1) is that α is enhanced almost up to the MG point by applying pressure. If so, the gap becomes about twice larger even with reduced δ as in Fig. 16(b). Since the abrupt enhancement of Δ is limited to the vicinity of the MG point as in Fig. 11(b), this fact may be an evidence of the strong frustration. Anyway, more detailed experiments and reliable theoretical knowledge on the relationship between the lattice structure and the model parameters are needed to settle this issue.

§5. Summary

Using the exact diagonalization and the recursion methods, we have investigated dynamical as well as static properties of the 1D J-J' model with coupling alternation, and compared with experiments of inorganic spin-Peierls compounds, $CuGeO_3$ and α'-NaV_2O_5. First, we have discussed the dynamical properties of the J-J' model without coupling alternation. The main points are:

(1) For $\alpha < \alpha_c$, the characteristics ([a]-[c] in §3.1 and §3.2) of the excitation spectrum are basically the same with those of the Heisenberg model.

(2) For $\alpha \sim \alpha_c$, the spectrum resembles that of the Haldane-Shastry model.

(3) As α further increases beyond α_c, there appear qualitatively different features from the Heisenberg model, summarized as [1]-[4] in §3.3.

Next, based on these results, we have studied a variety of features of the above SP compounds, particularly in view of the inelastic neutron scattering experiments. We itemize noticeable points below:

(4) Most characteristics of the complete neutron spectrum at 10K are quantitatively reproduced by the 1D J-J' model with $\alpha = 0.4$-0.45. Some experimental aspects cannot be explained, unless α is at least somewhat larger than α_c.

(5) Using $\alpha = 0.45$ (0.4), the strength of exchange coupling is evaluated at $J \sim 180$K, consistent with the estimations by other means. The dimerization parameter of the exchange coupling δ is found extremely small as ~ 0.001 (0.005).

(6) We have investigated the isolated branch appearing below the continuum or the double-gap structure for $CuGeO_3$ and α'-NaV_2O_5. The features of the second gap will be different between these two compounds.

(7) Pressure effect on $CuGeO_3$ is possibly interpreted as the increase of the frustration.

Our estimation of α is somewhat larger than the ones by $\chi(T)$, $\alpha \sim 0.35$.[64] Some causes of this discrepancy can be thought of. For example, evaluations are made at different temperatures between the two means. Our estimation does not allow for the temperature effect. There is quantitative ambiguity in neutron experiments at the present stage. Leaving aside such a quantitative point, we would like to emphasize that the effect of the large frustration beyond α_c is essential for various unique properties of $CuGeO_3$.

There remain many significant issues left for further studies. For example, although we treat a pure 1D model because the SP transition itself proves good one dimensionality, a relatively large magnetic dispersion perpendicular to the 1D chain was observed in $CuGeO_3$.[20] How do we interpret it? What effect does the large frustration have on the elemental substitutions? Furthermore, phonon softening has never been discovered in $CuGeO_3$,[12] which has always been a driving force of the organic SP transition so far. Does the large frustration shoulder such a role, as conjectured by Büchner *et al.* ?[65] To this end, the accumulation of experimental data on α'-NaV_2O_5 will be important.

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Appendix A: Single-Mode Approximation

In this Appendix, we summarize the properties of the single-mode approximation (SMA)[66, 30] in the J-J' model. As a kind of variational trial state for a spin-excited state with momentum q we consider

$$|\psi_q\rangle = S^z_q|0\rangle, \tag{A·1}$$

where $|0\rangle$ is the ground state—in this case exact. The excitation energy with respect to ψ_q is written as

$$E_q - E_0 = \frac{\langle\psi_q|\mathcal{H}|\psi_q\rangle}{\langle\psi_q|\psi_q\rangle} - E_0 = \frac{\langle 0|\left[S^z_{-q},\left[\mathcal{H},S^z_q\right]\right]/2|0\rangle}{\langle 0|S^z_{-q}S^z_q|0\rangle}$$
$$= \frac{K^{(1)}(q)}{S(q)} = \frac{\int d\omega\omega S(q,\omega)}{\int d\omega S(q,\omega)} \equiv \bar{\omega}_q, \tag{A·2}$$

where $K^{(1)}(q)$ is the first frequency moment. As seen in the last expression of eq.(A.2), $\bar{\omega}_q$ can be regarded as a kind of average of ω [the distribution function is $S(q,\omega)$], besides as a variational excitation energy. Hence the SMA becomes good when the distribution of $S(q,\omega)$ concentrates on a small range of ω; in particular if the weight of $S(q,\omega)$ is restricted to a certain sole value, the SMA becomes exact. We will prove that a pure single mode is realized for the exactly soluble cases at $q = \pi/2$ in the next Appendix. Inversely, when the distribution of $S(q)$ spreads over a wide range of ω evenly, the SMA is poor as the approximation of the lowest mode. In the meantime, since $\bar{\omega}_q$ is an average, it is an upper bound for the lowest spin excited mode. Therefore, one can use the SMA for a proof of the gaplessness.

For the Hamiltonian eq. (1.1), the first frequency moment is calculated as

$$K^{(1)}(q) = -2J\left[(1-\cos q)S_1 + \alpha(1-\cos 2q)S_2\right], \tag{A·3}$$

where S_i is the two point correlation function with respect to the ground state:

$$S_i = \frac{1}{N}\sum_j \langle S^z_j S^z_{j+i}\rangle. \tag{A·4}$$

Thus, by using the averages of the exact diagonalization, the SMA can be estimated. In Fig. 20 we depict the results for the J-J' model, thus obtained.

For $\alpha = 0$ and small q, the SMA curve is slightly higher than both of the two curves surrounding the TSC; this is due to the HOS processes as mentioned in §3.1.[27] For $\alpha = 0.2$ the result is close to that of the HS model, for which the SMA is estimated through the known analytical formulae of $K^{(1)}(q)$[67] and $S(q)$:[39]

$$\frac{\tilde{K}^{(1)}(q)}{J} = \frac{q}{16}(2\pi - q) + \frac{1}{8}(q-\pi)^2\ln\left(1-\frac{q}{\pi}\right), \tag{A·5}$$

$$\tilde{S}(q) = -\frac{1}{4}\ln\left(1-\frac{q}{\pi}\right). \tag{A·6}$$

This tendency has been discussed in §3.2.

For the MG case, $K^{(1)}(q) = J(1-\cos q)/4$ and $S(q) = (1-\cos q)/4$, therefore $\bar{\omega}_q = J$ independent of the value of q. As will be shown in the next Appendix, the spectrum becomes purely single at $q = \pi/2$; the SMA becomes exact then. Similarly, the SMA is relatively good for $\alpha = 0.45$ and $q \sim \pi/2$, though not exact. On the other hand, for $q = 0$ or π the SMA is poor as the lowest excitation. In the viewpoint of an average, however, the SMA indicates that there exists sizable contribution above $\omega = J$ to balance the lower excitation modes, as seen in Fig. 7(a).

To discuss the existence of the gap quantitatively in the SMA, larger system sizes are desired.

Appendix B: Proof of a Single Mode for Exactly Soluble Cases

In this Appendix, we describe a proof à la Majumdar and Ghosh[1] that $S(q,\omega)$ at $q = \pi/2$ in the exactly soluble cases ($2\alpha+\delta = 1$) is a pure single mode with a excitation energy independent of system size. The case of

$\delta = 0$ has been already discussed,[7] and a more compact proof was given of ψ_e being an eigenstate.[68]

If $S^z_q|\Psi_0\rangle$ becomes an eigenstate of $\mathcal{H}$, the surviving contribution to the sum of eq. (2.4) is given only by this eigenstate, due to the orthogonality of the eigenstates. Therefore, what we have to do for the proof is to show $S^z_q|\Psi_0\rangle$ is an eigenstate of $\mathcal{H}$, and then to estimate its eigenvalue.

The proof that $\psi_e \equiv S^z_{q=\pi/2}|\Psi_0\rangle$ becomes an eigenstate of $\mathcal{H}$ is obtained by an analogous treatment with the proof of the ground state.[1] Henceforth, we consider systems of $N = 4I$ (I: integer). Before going to the proof, we summarize notations and relations. Like a singlet pair $[i,j]$ of eq. (2.3), we denote a triplet pair by

$$\{i,j\} = \alpha(i)\beta(j) + \beta(i)\alpha(j). \tag{B·1}$$

Useful relations for this proof are as follows:[1]

$$\frac{1}{2}\left(1 - 4\boldsymbol{S}_\ell \cdot \boldsymbol{S}_m\right)[\ell,m] = 2[\ell,m], \tag{B·2}$$

$$\frac{1}{2}\left(1 - 4\boldsymbol{S}_\ell \cdot \boldsymbol{S}_m\right)\{\ell,m\} = 0, \tag{B·3}$$

$$\frac{1}{2}\left(1 - 4\boldsymbol{S}_\ell \cdot \boldsymbol{S}_m\right)[k,\ell][m,n] = [\ell,m][n,k], \tag{B·4}$$

$$\frac{1}{2}\left(1 - 4\boldsymbol{S}_\ell \cdot \boldsymbol{S}_m\right)[k,\ell]\{m,n\}$$
$$= \frac{1}{2}\left(1 - 4\boldsymbol{S}_\ell \cdot \boldsymbol{S}_m\right)\{k,\ell\}[m,n] = -[\ell,m]\{n,k\}, \tag{B·5}$$

$$[k,\ell][m,n] + [k,n][\ell,m] + [k,m][n,\ell] = 0, \tag{B·6}$$

$$[k,\ell]\{m,n\} + [\ell,m]\{n,k\} + [m,n]\{k,\ell\} + [n,k]\{\ell,m\} = 0. \tag{B·7}$$

First, we consider the case of $\delta = 0$. To make use of the above relations, we rewrite the Hamiltonian eq. (1.1) as,

$$\mathcal{H} = \frac{1}{4}NJ(1+\alpha) - \frac{1}{2}J\tilde{\mathcal{H}}, \tag{B·8}$$

with $\tilde{\mathcal{H}} = \tilde{\mathcal{H}}_1 + \tilde{\mathcal{H}}_2$ and,

$$\tilde{\mathcal{H}}_1 = \sum_{j=1}^{N}\frac{1}{2}(1 - 4\boldsymbol{S}_j \cdot \boldsymbol{S}_{j+1}), \tag{B·9}$$

$$\tilde{\mathcal{H}}_2 = \alpha\sum_{j=1}^{N}\frac{1}{2}(1 - 4\boldsymbol{S}_j \cdot \boldsymbol{S}_{j+2}). \tag{B·10}$$

As a ground-state eigenfunction, we take a form of eq. (2.2); substitution by ψ_2 makes no difference. Then, the function of our concern is given as,

$$\psi_e = S^z_{q=\frac{\pi}{2}}|\psi_1\rangle = \frac{1}{\sqrt{N}}\sum_{j=1}^{N}\exp\left(i\frac{\pi}{2}r_j\right)S^z_j|\psi_1\rangle$$
$$= \frac{1}{\sqrt{N}}\left(iS^z_1 - S^z_2 - iS^z_3 + S^z_4 + \cdots\right)[1,2]\cdots[N-1,N]$$
$$= \frac{1}{2\sqrt{N}}(1+i)\Big(\tau_1 - \tau_3 + \tau_5 - \tau_7 + \cdots$$
$$+ \tau_{N-3} - \tau_{N-1}\Big), \tag{B·11}$$

where

$$\tau_j = [1,2]\cdots[j-2,j-1]\{j,j+1\}[j+2,j+3]\cdots[N-1,N], \tag{B·12}$$

with odd j.

First of all, let us take up $\tilde{\mathcal{H}}\tau_1$, for example. Applying $\tilde{\mathcal{H}}_1$ and $\tilde{\mathcal{H}}_2$ to τ_1 and τ_3 and using eqs. (B.2)-(B.5), we obtain

$$\tilde{\mathcal{H}}_1\tau_1 = (N-2)\tau_1 - [2,3]\{4,1\}[5,6]\cdots[N-1,N]$$
$$+\{1,2\}[4,5][6,3]\cdots[N-1,N] + \cdots$$
$$+\{1,2\}[3,4]\cdots[N-2,N-1][N,N-3]$$
$$-[N,1]\{N-1,2\}[3,4]\cdots[N-3,N-2], \tag{B·13}$$

$$\tilde{\mathcal{H}}_1\tau_3 = (N-2)\tau_3 - [2,3]\{4,1\}[5,6]\cdots[N-1,N]$$
$$-[1,2][4,5]\{6,3\}[7,8]\cdots[N-1,N]$$
$$+[1,2]\{3,4\}[6,7][8,5][9,10]\cdots[N-1,N] + \cdots$$
$$+[1,2]\{3,4\}[5,6]\cdots[N-2,N-1][N,N-3], \tag{B·14}$$

$$\tilde{\mathcal{H}}_2\tau_1/\alpha = -[1,3]\{4,2\}[5,6]\cdots[N-1,N]$$
$$+[2,4]\{3,1\}[5,6]\cdots[N-1,N]$$
$$-2\{1,2\}[3,5][6,4][7,8]\cdots[N-1,N] - \cdots$$
$$-2\{1,2\}[3,4]\cdots[N-3,N-1][N,N-2]$$
$$+[N-1,1]\{N,2\}[3,4]\cdots[N-3,N-2]$$
$$-[N,2]\{N-1,1\}[3,4]\cdots[N-3,N-2], \tag{B·15}$$

$$\tilde{\mathcal{H}}_2\tau_3/\alpha = [1,3]\{4,2\}[5,6]\cdots[N-1,N]$$
$$-[2,4]\{3,1\}[5,6]\cdots[N-1,N]$$
$$-[1,2][3,5]\{6,4\}[7,8]\cdots[N-1,N]$$
$$+[1,2][4,6]\{5,3\}[7,8]\cdots[N-1,N]$$
$$-2[1,2]\{3,4\}[5,7][8,6][9,10]\cdots[N-3,N-2] - \cdots$$
$$-2[N-1,1][2,N]\{3,4\}[5,6]\cdots[N-3,N-2]. \tag{B·16}$$

To begin with, we consider terms appearing in $\tilde{\mathcal{H}}_2(\tau_1 - \tau_3)$. The first two terms in eqs.(B.15) and (B.16),

$$-2\left([1,3]\{4,2\} + [4,2]\{1,3\}\right)[5,6]\cdots[N-1,N],$$

are transformed into

$$2\left([3,4]\{2,1\} + [2,1]\{3,4\}\right)[5,6]\cdots[N-1,N], \tag{B·17}$$

by using eq. (B.7). We assign (or should say return) the first term of eq. (B.17) to $\tilde{\mathcal{H}}_2\tau_1$ instead of the first two terms of eq. (B.15), and the second term of eq. (B.17) to $\tilde{\mathcal{H}}_2\tau_3$ instead of the first two terms of eq. (B.16). Similarly, we can substitute

$$2[N-1,N]\{1,2\}[3,4]\cdots[N-3,N-2],$$

for the last two terms of eq. (B.15) by considering $\tilde{\mathcal{H}}_2(-\tau_{N-1} + \tau_1)$. Next, we apply the relation eq. (B.6) to the two singlet pairs on which $\tilde{\mathcal{H}}_2$ has been operated (namely pairs of irregular order) in residual terms of eq. (B.15). This substitution generates diagonal terms (τ_1) and the same kind of terms appearing in $\tilde{\mathcal{H}}_1\tau_1$ [from the

third term to the last but one in eq. (B.13)]. The residual off-diagonal terms, namely the second and the last ones in eq. (B.13) cancel out with the corresponding terms in eq. (B.14) and $\tilde{\mathcal{H}}_1\tau_{N-1}$, respectively. Consequently, eqs. (B.13) and (B.15) are modified as

$$(\tilde{\mathcal{H}}_1+\tilde{\mathcal{H}}_2)\tau_1=(N-2)\tau_1+\alpha N\tau_1 + (1-2\alpha)\Big(\{1,2\}[4,5][6,3][7,8]\cdots[N-1,N]+\cdots +\{1,2\}[3,4]\cdots[N-2,N-1][N,N-3]\Big) \quad \text{(B·18)}$$

If we put $\alpha=1/2$, the off-diagonal part in eq. (B.18) vanishes, resulting in

$$\tilde{\mathcal{H}}\tau_1=\left(\frac{3}{2}N-2\right)\tau_1. \quad \text{(B·19)}$$

As for $\tau_3,\tau_5,\cdots$, the same form with eq. (B.19) can be derived in the same manner. Then, by considering eqs. (B.11) and (B.8), the following eigenvalue equation can be obtained for the Majumdar-Ghosh model,

$$\mathcal{H}\psi_{\rm e}=\Big(-\frac{3}{8}N+1\Big)J\psi_{\rm e}. \quad \text{(B·20)}$$

Since the ground-state energy is $E_0=-3NJ/8$, the eigenvalue of $\psi_{\rm e}=S^z_{\pi/2}\psi_1$ is above E_0 by J, independent of the system size N.

This proof can be extended to the case of the exactly-soluble dimerized J-J' model eq. (2.1) with $2\alpha+\delta=1$. Note that ψ_2 cannot be chosen in eq. (B.11) here. Then, the eigenvalue equation corresponding to eq. (B.20) becomes

$$\mathcal{H}\psi_{\rm e}=\left[-\frac{3}{4}(1-\alpha)N+2(1-\alpha)\right]J\psi_{\rm e}. \quad \text{(B·21)}$$

Since the first term is the ground-state energy in this case,[3)] the energy increment is $2(1-\alpha)J$.

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Fig. 1. $S(q,\omega)$ of the Heisenberg model for $N = 26$. The poles are situated at the center of the spheres. The intensity of each pole is proportional to the *volume* of the sphere to emphasize the poles with weak intensity. The boundaries of the two-spinon continuum, $\omega_\ell(q)$ and $\omega_{\rm u}(q)$, are shown by solid lines. The lower edge of the higher-order-spinon continuum is indicated by a dashed line ($N = 26$), which is strongly dependent on N. The lowest quintet level for each q ($N = 14$) is marked with solid diamond.

Fig. 2. (a) Dependence on system size of residues which belong to the lowest two two-spinon branches for the Heisenberg model. The lowest (second-lowest) branch is indicated by solid symbols and 1' (open symbols and 2'). (b) Dependence on N of the poles belonging to the lowest four two-spinon branches (1'-4') for $\alpha = 0$. Symbols have the same meanings with (a). The inset shows the dependence on N of poles at $q = \pi$ as a function of $1/N^2$.

Fig. 3. (a) Total weight of residues outside the main continuum versus q/π for some values of $\alpha(\leq \alpha_{\rm c})$ and N. The poles by higher-order processes are located outside of the TSC for the present values of q (see Fig. 1). (b) Maximum values of the same quantity as a function of α for $0 \leq q/\pi \leq 0.5$. The arrows near the vertical axes indicate the direction of the dependence on N to the thermodynamic limit around their positions.

Fig. 4. $S(q,\omega)$ for $\alpha = 0.2$ and $N = 26$. The intensity of each pole is proportional to the *volume* of the sphere, and in the same scale with Fig. 1. The boundaries of the two-spinon continuum for the HS model, $\tilde{\omega}_\ell(q)$ and $\tilde{\omega}_{\rm u}(q)$, are shown by solid lines.

Fig. 5. (a) Static susceptibility as a function of q for $\alpha = 0, 0.2, 0.25, 0.4, 0.45, 0.5$ from bottom to top, respectively. Lines (except solid line) indicate polynomial fit of 26-site data as a guide for the eye. The solid line is the value for the HS model given by eq. (3.6). The arrow and cross on the vertical axis indicate the exact value both for the Heisenberg model and the HS model: $1/2\pi^2$. The inset is a further magnification of small-q regime to recognize singular behavior of the Heisenberg model. (b) Magnified figure of (a) for small q. Symbols are common in (a) and (b).

Fig. 6. (a) $K^{(2)}(q)$ and (b) $K^{(3)}(q)$ for some values of α. Symbols are common between (a) and (b). Lines (except solid line) are guide for the eye. The solid lines in both panels are of the HS model given by eqs. (3.7) and (3.8), respectively.

Fig. 7. (a) Residues for the MG model. The system with $N = 24$ is shown to include the wave number $\pi/2$. (b) $S(q, \omega)$ for the MG model ($N = 26$). In both (a) and (b), both contributions from Ψ_+ and Ψ_- are plotted together, and the weight is indicated by the *area* of the circles.

Fig. 8. (a) Dependence on N of residues for the first (1') and second (2') lowest dominant branches as a function of q. (b) The same for the third (3') and fourth (4') dominant branches. Both contributions from Ψ_+ and Ψ_- are plotted together. (c) Dependence on N of the pole positions corresponding to the residues in (a) and (b). The cross and arrow on the right vertical axis show the extrapolated pole position of the branch 1' in the thermodynamic limit, which means the gap of the MG model: $\Delta/J \sim 0.233$ in our estimation by the fourth-order polynomial fit. The inset shows the movement of the poles at $q = \pi$ as the system size changes. Since the spectrum is symmetric with respect to $q = \pi/2$, we only show $0.5 \leq q/\pi \leq 1.0$.

Fig. 9. Residues at $q = \pi$ as a function of ω/J for some system sizes. The connected points with numbers (1' etc.) correspond to the poles in Fig. 8. The arrow indicates the peak near the upper edge of the main continuum. Since the number of coefficients used in the recursion method is different for different systems, namely 30 for $N \geq 22$ and 100 for $N \leq 20$, many poles with weak intensity appear for the small systems.

Fig. 10. $S(q, \omega)$ of $N = 26$ for (a) $\alpha = 0.35$, (b) 0.4 and (c) 0.45. In this size the ground state is $Q = \pi$. The intensity is proportional to the *area* of the circle, and in the same scale with Fig. 7(b).

Fig. 11. (a) The ground-state energy, (b) the lowest excited energy at $q = \pi/2$ and the gap, each as a function of α. Each value is extrapolated from the finite-size data (N = 8-26) by using some orders of polynomial fit. The values for the HS model are indicated by crosses and arrows on the vertical axes. The energy and the gap were previously estimated in some references.[4)]

Fig. 12. $S(q)$ for some large values of α. Each line connects the data of $N = 26$ except for $\alpha = 0.5$. The arrows along the right vertical axis indicate the directions of the dependence on N of $S(\pi)$ for $N \to \infty$.

Fig. 13. Comparison of $S(q)$ between the diagonalization results for $\delta = 0$ and the experimental results for 10K and 20K, which are taken from Fig. 2 of ref. 19. The arrows along the right vertical axis indicate the directions of the dependence on N for $N \to \infty$.

Fig. 14. $S(q, \omega)$ at a fixed value of $\delta = 0.05$ for three values of α, (a) 0, (b) 0.2 and (c) 0.4. $N = 26$. In each figure, the intensity is proportional to the *area* of the circle, and in the same scale with Figs. 7(b) and 10.

Fig. 15. Lowest excitation at $q = \pi/2$ as a function of δ for some values of α. Shown data are extrapolated from finite-size results of N = 8-24 with using eq. (2.9) and $m = 1$. The values for $\delta = 0$ correspond to Fig. 11(b). The arrow on the vertical axis indicates the value of des Cloizeaux-Pearson, $\pi/2$. Shaded lines are guide for the eye. The dash-dotted line shows the exactly soluble case, as given in Appendix B.

Fig. 16. (a) Gap of the model eq. (2.1) as a function of δ for some values of α. Each value is extrapolated from the results of N = 8-26 by using eq. (2.9). The arrow on the vertical axis indicates the gap for the MG model. If we fit for each α with power functions, the resultant powers are 0.71 ($\alpha = 0.2$), 0.61 (0.35), 0.58 (0.4) and 0.51 (0.45). Dashed (dash-dotted) line indicates a plausible value of α'-NaV_2O_5 for $\Delta = 85$K and $J = 441$K[51)] ($\Delta = 98$K and $J = 560$K[49)]). The data for small δ and/or α are omitted, because reliable results cannot be obtained by extrapolations. Size of the symbols and width of the lines roughly represent the inaccuracy of the extrapolation. (b) Magnification of (a) for a small-δ regime. Dash-dotted (dashed) line indicates a value of $CuGeO_3$ ($\Delta = 24$K) for $J = 180$K (160K). See text.

Fig. 17. Total energy of the model eq. (2.1) as a function of δ for some values of α. Each value is extrapolated from the results of N = 8-26 by using eq. (2.9). If we fit the data for each α with power functions in this range of δ, the resultant powers are 1.42 ($\alpha = 0$), 1.30 (0.2), 1.13 (0.35), 1.07 (0.4) and 1.03 (0.45). Dash-dotted line indicates the soluble case of $2\alpha + \delta = 1$.

Fig. 18. (a) Dependence on system size of the pole positions of the lowest two branches. The parameters roughly correspond to $CuGeO_3$. The subtle asymmetry with respect to $q = \pi/2$ is due to numerical round-off errors. The inset shows the dependence on N at $q = \pi$ for N = 8-26, fitted by eq. (2.9). (b) Dependence on N of the corresponding residues. The arrow on the right axis indicates the extrapolated value for $N \to \infty$ of the branch 1'.

Fig. 19. (a) Dependence on N of the pole positions for the lowest three *dominant* branches. The parameters roughly correspond to α'-NaV_2O_5. Concerning the branch 3', we show only for $q/\pi \gtrsim 0.4$. By '+' we represent poles with tiny weight; to avoid mess we plot only for $q/\pi \gtrsim 0.6$ and under the branch 3'. The inset shows the dependence on N at $q = \pi$ for N = 8-26 with polynomial fit. (b) Dependence on N of the corresponding residues.

Fig. 20. Lowest spin excited modes for the 1D J-J' model obtained by the SMA. For the MG case (dash-dotted line), $\omega = J$. Otherwise, the diagonalization results are used. Dotted lines are guide for the eye. For comparison for $\alpha = 0$, the exact lower (dCP) and upper edges of the TSC are shown with dashed lines. The lower edge for $\alpha = 0.45$ obtained by the recursion method (N = 20-26) is indicated by cross and plus. The SMA and the lowest edge for the HS model is indicated by solid and long dash-dotted lines, respectively. The extrapolated values of the gap (exact) are shown by arrows on the vertical axis for three values of α.